

REC'D 02 OCT 2000

WIPO

PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference R1331-PCT	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP99/03599	International filing date (day/month/year) 26/05/1999	Priority date (day/month/year) 26/05/1998
International Patent Classification (IPC) or national classification and IPC C23C4/10		
Applicant UNIVERSITEIT GENT et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 4 sheets, including this cover sheet.

- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 3 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☒ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand 20/12/1999	Date of completion of this report 29.09.2000
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Schmidt, O  Telephone No. +49 89 2399 8438 

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP99/03599

**I. Basis of the report**

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

**Description, pages:**

1-19 as originally filed

**Claims, No.:**

1-17 as received on 21/08/2000 with letter of 14/08/2000

**Drawings, sheets:**

1/3-3/3 as originally filed

2. The amendments have resulted in the cancellation of:

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

**IV. Lack of unity of invention**

1. In response to the invitation to restrict or pay additional fees the applicant has:

- ☒ restricted the claims.  
☐ paid additional fees.  
☐ paid additional fees under protest.  
☐ neither restricted nor paid additional fees.

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP99/03599

2. ☐ This Authority found that the requirement of unity of invention is not complied and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.
3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is
- ☒ complied with.
- ☐ not complied with for the following reasons:
4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:
- ☒ all parts.
- ☐ the parts relating to claims Nos. .

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes: Claims 1-17
	No: Claims
Inventive step (IS)	Yes: Claims 1-17
	No: Claims
Industrial applicability (IA)	Yes: Claims 1-17
	No: Claims

2. Citations and explanations

**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

**see separate sheet**

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

---

International application No. PCT/EP99/03599

**SECTION V.:**

1.

1.1 The following documents D1 and D3 are referred to in this report:

D1: US-a-5 196 400 (CHEN CHIOU T ET AL) 23 March 1993 (1993-03-23) cited in the application;

D3: CH 648 358 a (CASTOLIN SA) 15 March 1985 (1985-03-15)

1.2 The new set of claims filed with the letter dated 14.8.00 meet the requirements of Articles 6 and 34(2)(b) PCT.

1.3 The objective of the application is to provide a method of depositing by flame or plasma spraying at atmospheric pressure a metal oxide containing layer onto a substrate having a thickness of at least 5 mm, comprising the step of depositing an additional noble metal with the coating to increase thermal conductivity of the coating. Another object of the present application is to provide a composite comprising a substrate and a coating deposited thereon in accordance with the aforementioned method. This objective is solved by the subject-matter according to claims 1 to 17. Document D1 and D3 do not disclose a flame or plasma sprayed coated substrate of at least 5 mm thickness and comprising the addition of a noble metal to increase thermal conductivity of the coating nor a method of preparing said composite.

The subject-matter of claims 1 to 17 meet the requirements of Article 33(2)(3)(4) PCT.

**SECTION VIII.:**

2. The general statement in the description on page 19, last paragraph which implies that the extent of protection may be expanded in some vague and not precisely defined way is not allowable (cf. the PCT Guidelines, C-III, 4.3a). Moreover, the description would need to be adapted to the new revised set of claims in the regional phase of the application later on.

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

Bird, Ariane  
Bird Goen & Co.  
Vilvoordsebaan 92  
3020 Winksele  
BELGIQUE

RECEIVED

06 OKT. 2000

PCT

NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT  
(PCT Rule 71.1)

Date of mailing  
(day/month/year) 29.09.2000

Applicant's or agent's file reference  
R1331-PCT

**IMPORTANT NOTIFICATION**

International application No.  
PCT/EP99/03599

International filing date (day/month/year)  
26/05/1999

Priority date (day/month/year)  
26/05/1998

Applicant  
UNIVERSITEIT GENT et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

**4. REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/



European Patent Office  
D-80298 Munich  
Tel. +49 89 2399 - 0 Tx: 523656 epmu d  
Fax: +49 89 2399 - 4465

Authorized officer

Koutsoftas, P

Tel. +49 89 2399-7273



# PATENT COOPERATION TREATY

## PCT

### INTERNATIONAL PRELIMINARY EXAMINATION REPORT


(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>R1331-PCT</b>		<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
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Applicant <b>UNIVERSITEIT GENT et al.</b>			

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- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand <b>20/12/1999</b>	Date of completion of this report <b>29.09.2000</b>
Name and mailing address of the international preliminary examining authority:   <b>European Patent Office</b> <b>D-80298 Munich</b> <b>Tel. +49 89 2399 - 0 Tx: 523656 epmu d</b> <b>Fax: +49 89 2399 - 4465</b>	Authorized officer  <b>Schmidt, O</b>  <b>Telephone No. +49 89 2399 8438</b>



**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP99/03599

**I. Basis of the report**

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- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

**IV. Lack of unity of invention**

1. In response to the invitation to restrict or pay additional fees the applicant has:

- ☒ restricted the claims.  
☐ paid additional fees.  
☐ paid additional fees under protest.  
☐ neither restricted nor paid additional fees.

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP99/03599

2. ☐ This Authority found that the requirement of unity of invention is not complied and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.
3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is
- ☒ complied with.
- ☐ not complied with for the following reasons:
4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:
- ☒ all parts.
- ☐ the parts relating to claims Nos. .

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes: Claims 1-17
	No: Claims
Inventive step (IS)	Yes: Claims 1-17
	No: Claims
Industrial applicability (IA)	Yes: Claims 1-17
	No: Claims

**2. Citations and explanations**

see separate sheet

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet



**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/EP99/03599

**SECTION V.:**

1.

1.1 The following documents D1 and D3 are referred to in this report:

D1: US-a-5 196 400 (CHEN CHIOU T ET AL) 23 March 1993 (1993-03-23) cited in the application;

D3: CH 648 358 a (CASTOLIN SA) 15 March 1985 (1985-03-15)

1.2 The new set of claims filed with the letter dated 14.8.00 meet the requirements of Articles 6 and 34(2)(b) PCT.

1.3 The objective of the application is to provide a method of depositing by flame or plasma spraying at atmospheric pressure a metal oxide containing layer onto a substrate having a thickness of at least 5 mm, comprising the step of depositing an additional noble metal with the coating to increase thermal conductivity of the coating. Another object of the present application is to provide a composite comprising a substrate and a coating deposited thereon in accordance with the aforementioned method. This objective is solved by the subject-matter according to claims 1 to 17. Document D1 and D3 do not disclose a flame or plasma sprayed coated substrate of at least 5 mm thickness and comprising the addition of a noble metal to increase thermal conductivity of the coating nor a method of preparing said composite.

The subject-matter of claims 1 to 17 meet the requirements of Article 33(2)(3)(4) PCT.

**SECTION VIII.:**

2. The general statement in the description on page 19, last paragraph which implies that the extent of protection may be expanded in some vague and not precisely defined way is not allowable (cf. the PCT Guidelines, C-III, 4.3a). Moreover, the description would need to be adapted to the new revised set of claims in the regional phase of the application later on.

## CLAIMS

1. A composite comprising: a substrate and a coating deposited on said substrate, the coating being deposited by **flame or plasma spraying at atmospheric pressure**, the thickness of the coating being at least 5 mm, more preferably greater than 8mm, the coating comprising metal oxides **and the deposited coating comprising the addition of a noble metal to increase thermal conductivity of the coating.**
2. A composite comprising: a substrate and a coating deposited on said substrate, the coating being deposited by **flame or plasma spraying at atmospheric pressure**, the thickness of the coating being at least 5 mm, more preferably greater than 8mm, the coating comprising a superconductor precursor and at least 10% of the coating is in a superconductive phase **as deposited.**
3. The composite according to claim 1, wherein the coating is a superconductive precursor and least 10% of the coating is in a superconductive phase **as deposited.**
4. The composite according to claim 1, wherein the noble metal is silver.
5. The composite according to claim 4, wherein up to 30% silver is included in the metal oxides.
6. The composite according to any of claims 1 to 5, wherein the composite is a target for a sputtering magnetron.
7. The composite according to claim 6, wherein the target is cylindrical.
8. The composite in accordance with any previous claim, wherein the coating has

a thermal conductivity of between 1 and 5  $\text{Wm}^{-1}\text{K}^{-1}$ .

9. The composite in accordance with any previous claim, wherein the thermal conductivity of the composite or the target through the substrate and the coating  
5 is in the range 25 to 125  $\text{Wm}^{-1}\text{K}^{-1}$ .

10. The composite in accordance with any previous claim, wherein the coating has an electrical resistivity of lower than  $15 \times 10^{-6} \text{ Ohm.m}$ , more preferably lower than  $10 \times 10^{-6}$  and most preferably less than  $5 \times 10^{-6} \text{ Ohm.m}$ .

10  
11. A method of depositing by **flame or plasma spraying at atmospheric pressure** a layer onto a substrate, the layer having a thickness of at least 5 mm, more preferably greater than 8mm, the coating comprising metal oxides, the method including the step of depositing an additional noble metal with the  
15 coating to increase thermal conductivity of the coating.

12. A method of depositing by **flame or plasma spraying at atmospheric pressure** a layer onto a substrate, the layer having a thickness of at least 5 mm, more preferably greater than 8mm, the coating comprising a superconductive precursor and at least 10% of the layer being in a superconductive phase as  
20 deposited.

13. The method according to claim 11, wherein the noble metal is silver.

25 14. The method according to claim 13, wherein up to 30% silver is included in the material to be sprayed.

15. The method according to any of claims 11 to 14, wherein the spraying step includes spraying a material through a spraying head, the material being in the

form of a powder, a slurry or a solution.

16. The method according to any of claims 11 to 15, further including the step of cooling the substrate.

17. The method according to claim 16, wherein the cooling is with a cryogenic liquid.

**PCT**

From the INTERNATIONAL BUREAU

**NOTIFICATION OF THE RECORDING  
 OF A CHANGE**

(PCT Rule 92bis.1 and  
 Administrative Instructions, Section 422)

To:

BIRD, William  
 Bird Goën & Co.  
 Vilvoordsebaan 92  
 B-3020 Winksele  
 BELGIQUE

Date of mailing (day/month/year) 22 September 2000 (22.09.00)	<b>IMPORTANT NOTIFICATION</b>
Applicant's or agent's file reference R1331-PCT	
International application No. PCT/EP99/03599	International filing date (day/month/year) 26 May 1999 (26.05.99)

1. The following indications appeared on record concerning:

☐ the applicant ☐ the inventor ☒ the agent ☐ the common representative

Name and Address

BIRD, William  
 Bird Goën & Co.  
 Termerestraat 1  
 B-3020 Winksele  
 Belgium

State of Nationality

State of Residence

Telephone No.

32 16 48 05 62

Facsimile No.

32 16 48 05 28

Teleprinter No.

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:

☐ the person ☐ the name ☒ the address ☐ the nationality ☐ the residence

Name and Address

BIRD, William  
 Bird Goën & Co.  
 Vilvoordsebaan 92  
 B-3020 Winksele  
 Belgium

State of Nationality

State of Residence

Telephone No.

32 16 48 05 62

Facsimile No.

32 16 48 05 28

Teleprinter No.

3. Further observations, if necessary:

4. A copy of this notification has been sent to:

☒ the receiving Office ☐ the designated Offices concerned  
☐ the International Searching Authority ☒ the elected Offices concerned  
☒ the International Preliminary Examining Authority ☐ other:

The International Bureau of WIPO  
 34, chemin des Colombettes  
 1211 Geneva 20, Switzerland

Authorized officer

C. Cupello

Facsimile No.: (41-22) 740.14.35

Telephone No.: (41-22) 338.83.38

## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents  
 United States Patent and Trademark  
 Office  
 Box PCT  
 Washington, D.C.20231  
 ÉTATS-UNIS D'AMÉRIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 11 February 2000 (11.02.00)	
International application No. PCT/EP99/03599	Applicant's or agent's file reference R1331-PCT
International filing date (day/month/year) 26 May 1999 (26.05.99)	Priority date (day/month/year) 26 May 1998 (26.05.98)
Applicant HOSTE, Serge et al	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
 20 December 1999 (20.12.99)

☐ in a notice effecting later election filed with the International Bureau on:  
 \_\_\_\_\_

2. The election ☒ was  
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer R. E. Stoffel Telephone No.: (41-22) 338.83.38
---	---

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>R1331-PCT</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/EP 99/ 03599</b>	International filing date (day/month/year) <b>26/05/1999</b>	(Earliest) Priority Date (day/month/year) <b>26/05/1998</b>
Applicant <b>UNIVERSITEIT GENT et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 6 sheets.



it is also accompanied by a copy of each prior art document cited in this report.

**1. Basis of the report**

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.



the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing:



contained in the international application in written form.



filed together with the international application in computer readable form.



furnished subsequently to this Authority in written form.



furnished subsequently to this Authority in computer readable form.



the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.



the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☒ **Certain claims were found unsearchable** (See Box I).

3. ☒ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,



the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

**SPRAYING METHOD TO FORM A THICK COATING AND PRODUCTS OBTAINED**

5. With regard to the **abstract**,



the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.



as suggested by the applicant.



because the applicant failed to suggest a figure.



because this figure better characterizes the invention.

1



None of the figures.

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/EP 99/ 03599

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2. ☒ Claims Nos.: 9  
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:  
**SEE FURTHER INFORMATION SHEET**
  
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

**SEE FURTHER INFORMATION SHEET**

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
  
2. ☒ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
  
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
  
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.



## FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1-8,10-14

Claims for a composite comprising an oxide-containing coating deposited by spraying and having a certain minimum thickness, a sputter target having a layer of a superconductor precursor, also deposited by spraying and having a minimum thickness, and methods for producing each of these products, respectively.

2. Claims: 15,16

Reconditioned sputter target and method for its manufacture by flame or plasma spraying material into the sputtered erosion groove.

The only common concept linking subjects 1 and 2 is the application of spraying for depositing layers of material. Since this feature is known from many prior art documents (see for example search report) there is effectively no common concept to connect the two groups of claims. Furthermore, while subject 1 addresses the problem of producing relatively thick deposits of oxides by means of spraying, subject 2 solves the unrelated problem of spraying an unspecified material to repair local damage on a target.

Therefore there is a lack of unity between the two subjects.

Nevertheless, since the EPO has already searched the subject-matter of claims 15 and 16 in connection with the priority application EP98870120, an International search report is issued for both subjects.

## FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box I.2

Claims Nos.: 9

Claim 9 relates to a product defined (inter alia) by reference to the following parameter:

the result of a test involving impact of a ball consisting of an unspecified steel, wherein both the weight of the ball and the height from which it is dropped are arbitrarily chosen.

The use of this parameter in the present context is considered to lead to a lack of clarity within the meaning of Article 6 PCT. It is impossible to compare the parameter the applicant has chosen to employ with what is set out in the prior art. The lack of clarity is such as to render a meaningful complete search impossible. Consequently, the search has been restricted to the remaining claims

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/03599

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C23C4/10 C23C4/12 C23C14/34

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C23C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 196 400 A (CHEN CHIOU T ET AL) 23 March 1993 (1993-03-23) cited in the application column 3, line 52 - column 4, line 16 ---	1-5, 10-14
X	EP 0 330 196 A (PERKIN ELMER CORP) 30 August 1989 (1989-08-30) page 5, line 20 - line 25; claims 1,8 ---	1,2,10, 12-14
X	CH 648 358 A (CASTOLIN SA) 15 March 1985 (1985-03-15) claim 1 ---	1,10, 12-14
X	DD 277 471 A (MANSFELD KOMBINAT W PIECK VEB) 4 April 1990 (1990-04-04) page 2, line 21 - line 23 ---	15,16
	--- -/--	

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&amp;" document member of the same patent family

Date of the actual completion of the international search

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Date of mailing of the international search report

16/09/1999

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Patterson, A

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/03599

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE 40 15 387 A (LEYBOLD AG) 21 November 1991 (1991-11-21) the whole document	15, 16
A	----- EP 0 377 073 A (MITSUBISHI METAL CORP) 11 July 1990 (1990-07-11) page 3, line 41 - line 57; table 2 page 7, line 7 - line 23; table 4 -----	5-8

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/03599

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 5196400	A	23-03-1993	NONE	
EP 0330196	A	30-08-1989	CN 1036286 A JP 1309952 A	11-10-1989 14-12-1989
CH 648358	A	15-03-1985	NONE	
DD 277471	A	04-04-1990	NONE	
DE 4015387	A	21-11-1991	NONE	
EP 0377073	A	11-07-1990	JP 2156079 A JP 2157150 A KR 9507087 B US 5077269 A KR 9507088 B	15-06-1990 15-06-1990 30-06-1995 31-12-1991 30-06-1995

## CLAIMS

1. A composite comprising: a substrate and a coating deposited on said substrate, the coating being deposited by spraying, the thickness of the coating being at least 5 mm, more preferably greater than 8mm, the coating comprising metal oxides.
2. The composite according to claim 1, wherein the coating comprises a superconductor precursor and at least 10% of the coating is in a superconductive phase
3. The composite according to claim 1 or 2, wherein the composite is a target for a sputtering magnetron.
4. The composite according to claim 3, wherein the target is cylindrical.
5. The target for a sputtering magnetron comprising: a cylindrical substrate and a coating deposited on said substrate, the coating being deposited by spraying, the thickness of the coating being at least 3mm, more preferably at least 5 mm, most preferably greater than 8mm, the coating comprising a superconductor precursor and at least 10% of the coating is in a superconductive phase.
6. The target or a composite in accordance with any previous claim, wherein the coating has a thermal conductivity of between 1 and 5  $\text{Wm}^{-1}\text{K}^{-1}$ .
7. The target or a composite in accordance with any previous claim, wherein the thermal conductivity of the composite or the target through the substrate and the coating is in the range 25 to 125  $\text{Wm}^{-1}\text{K}^{-1}$ .
8. The target or a composite in accordance with any previous claim, wherein the coating has an electrical resistivity of lower than  $15 \times 10^{-6} \text{ Ohm.m}$ , more preferably lower than  $10 \times 10^{-6}$  and most preferably less than  $5 \times 10^{-6} \text{ Ohm.m}$ .

9. The target or a composite in accordance with any previous claim, wherein the coating can withstand impact of a 0.036 kg steel ball from a height of 1 metre, preferably from 1.5 metre.

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10. The target or a composite in accordance with any previous claim, wherein the spraying is one of plasma spraying and flame spraying.

11. A method of depositing by spraying a superconductor precursor layer onto a cylindrical target for a sputtering magnetron, the layer having a thickness of at least 3 mm, and at least 10% of the layer being in a superconductive phase.

12. A method of depositing by spraying a layer onto a substrate, the layer having a thickness of at least 5 mm, and the coating comprising metal oxides.

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13. The method according to claim 11 or 12, wherein the spraying step is one of flame spraying and low-pressure or atmospheric pressure plasma spraying.

14. The method according to claim 13, wherein the spraying step includes spraying a material through a spraying head, the material being in the form of a powder, a slurry or a solution.

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15. A method of reconditioning a used target for a sputtering magnetron having an erosion groove in the target material, comprising the step of: flame or atmospheric pressure plasma spraying target material into the erosion groove.

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16. A reconditioned target for a sputtering magnetron, comprising:  
an erosion groove in the target material; and target material flame sprayed or atmospheric plasma sprayed into said groove to restore the thickness of the target material to that of the unused material.

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C23C 4/10, 4/12, 14/34

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(71) Applicant (for all designated States except US): UNIVER-  
SITEIT GENT [BE/BE]; St. Pietersnieuwstraat 25, B-9000  
Gent (BE).

**(72) Inventors; and**

(75) **Inventors/Applicants (for US only):** HOSTE, Serge [BE/BE]; Gaverse steenweg 579, B-9280 Merelbeke (BE). PERSYN, Frans [BE/BE]; Oude boongaardlaan 23, B-9080 Lochrist (BE). VAN DRIESCHE, Isabel [BE/BE]; Wareslagstraat 94, B-9250 Waasmunster (BE).

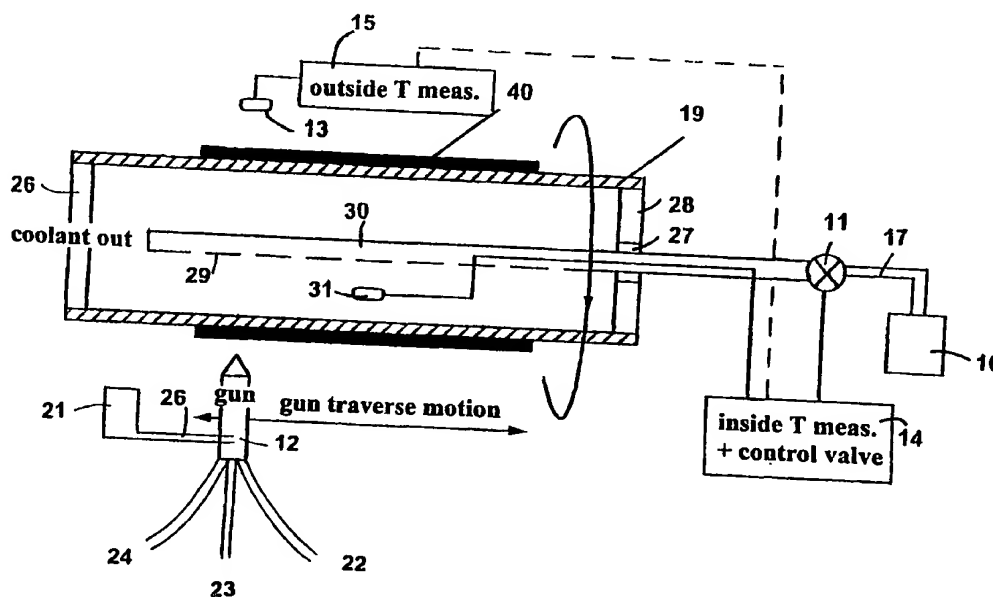
(74) Agents: BIRD, William et al.; Bird Goën & Co., Termerestraat  
1, B-3020 Winksele (BE).

(81) Designated States: JP, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

**Published**

*With international search report.*

(54) Title: SPRAYING METHOD TO FORM A THICK COATING AND PRODUCTS OBTAINED



**(57) Abstract**

A method and an apparatus for spraying materials onto a substrate to produce a coating thereon is described which allows very thick layers of complex metal oxides to be produced. The apparatus and method are particularly suitable for producing superconducting coatings.



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# INTERNATIONAL SEARCH REPORT

International Application No

PC/EP 99/03599

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C23C4/10 C23C4/12 C23C14/34

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 C23C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 196 400 A (CHEN CHIOU T ET AL) 23 March 1993 (1993-03-23) cited in the application column 3, line 52 - column 4, line 16 ---	1-5, 10-14
X	EP 0 330 196 A (PERKIN ELMER CORP) 30 August 1989 (1989-08-30) page 5, line 20 - line 25; claims 1,8 ---	1,2,10, 12-14
X	CH 648 358 A (CASTOLIN SA) 15 March 1985 (1985-03-15) claim 1 ---	1,10, 12-14
X	DD 277 471 A (MANSFELD KOMBINAT W PIECK VEB) 4 April 1990 (1990-04-04) page 2, line 21 - line 23 ---	15,16
	-/--	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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- "A" document defining the general state of the art which is not considered to be of particular relevance
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Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Patterson, A

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/03599

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# INTERNATIONAL SEARCH REPORT

International application No.

PCT/EP 99/03599

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☒ Claims Nos.: 9  
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:  
**SEE FURTHER INFORMATION SHEET**
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

**SEE FURTHER INFORMATION SHEET**

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☒ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

# INTERNATIONAL SEARCH REPORT

International Application No. PCT/EP 99 03599

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1-8,10-14

Claims for a composite comprising an oxide-containing coating deposited by spraying and having a certain minimum thickness, a sputter target having a layer of a superconductor precursor, also deposited by spraying and having a minimum thickness, and methods for producing each of these products, respectively.

2. Claims: 15,16

Reconditioned sputter target and method for its manufacture by flame or plasma spraying material into the sputtered erosion groove.

The only common concept linking subjects 1 and 2 is the application of spraying for depositing layers of material. Since this feature is known from many prior art documents (see for example search report) there is effectively no common concept to connect the two groups of claims. Furthermore, while subject 1 addresses the problem of producing relatively thick deposits of oxides by means of spraying, subject 2 solves the unrelated problem of spraying an unspecified material to repair local damage on a target.

Therefore there is a lack of unity between the two subjects.

Nevertheless, since the EPO has already searched the subject-matter of claims 15 and 16 in connection with the priority application EP98870120, an International search report is issued for both subjects.

# INTERNATIONAL SEARCH REPORT

International Application No. PCT/EP 99 03599

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box I.2

Claims Nos.: 9

Claim 9 relates to a product defined (inter alia) by reference to the following parameter:  
the result of a test involving impact of a ball consisting of an unspecified steel, wherein both the weight of the ball and the height from which it is dropped are arbitrarily chosen.  
The use of this parameter in the present context is considered to lead to a lack of clarity within the meaning of Article 6 PCT. It is impossible to compare the parameter the applicant has chosen to employ with what is set out in the prior art. The lack of clarity is such as to render a meaningful complete search impossible. Consequently, the search has been restricted to the remaining claims

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/03599

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 5196400	A	23-03-1993	NONE	
EP 0330196	A	30-08-1989	CN 1036286 A JP 1309952 A	11-10-1989 14-12-1989
CH 648358	A	15-03-1985	NONE	
DD 277471	A	04-04-1990	NONE	
DE 4015387	A	21-11-1991	NONE	
EP 0377073	A	11-07-1990	JP 2156079 A JP 2157150 A KR 9507087 B US 5077269 A KR 9507088 B	15-06-1990 15-06-1990 30-06-1995 31-12-1991 30-06-1995